



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : A61L 9/04, B65D 75/30	A1	(11) International Publication Number: WO 99/47182 (43) International Publication Date: 23 September 1999 (23.09.99)
(21) International Application Number: PCT/EP99/01789 (22) International Filing Date: 16 March 1999 (16.03.99) (30) Priority Data: 60/078,562 19 March 1998 (19.03.98) US (71) Applicant (for AE AU BB CA CY GB GD GH GM IE IL KE LC LK LS MN MW NZ SD SG SL SZ TT UG ZA ZW only): UNILEVER PLC (GB/GB); Unilever House, Blackfriars, London EC4P 4BQ (GB). (71) Applicant (for all designated States except AE AU BB CA CY GB GD GH GM IE IL IN KE LC LK LS MN MW NZ SD SG SL SZ TT UG ZA ZW): UNILEVER N.V. [NL/NL]; Weena 455, NL-3013 AL Rotterdam (NL). (71) Applicants (for IN only): HINDUSTAN LEVER LIMITED [IN/IN]; Hindustan Lever House, 165/166 Backbay Reclamation, Mumbai 400 020 (IN). INTERNATIONAL FLAVORS & FRAGRANCES INC. [US/US]; 521 West 57th Street, New York, NY 10019 (US).		(72) Inventors: WARREN, Craig; 316 Fairfield Avenue, Long Branch, County of Monmouth, NJ 07740 (US). RAMS-BOTHAM, John; Veenwal #1, NL-3871 KE Hoevelaken (NL). BELT, Marcel; Lever, Divisione Lever, Via Nino Bonnet, 6/A, I-20154 Milan (IT). (74) Agent: MOLE, Peter, Geoffrey; Unilever PLC, Patent Dept., Colworth House, Sharnbrook, Bedford MK44 1LQ (GB). (81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i>
(54) Title: TIME RELEASE FRAGRANCE SACHET, METHOD OF USING SAME AND METHOD OF FABRICATING SAME <div data-bbox="496 1144 1096 1682" data-label="Image"> </div> (57) Abstract <p>Described is a time release fragrance sachet for air freshening, particularly for use in clothing storage cabinets. The sachets comprise a container fabricated from a substance either sufficiently porous for perfumes to pass therethrough or having openings capable of permitting fragrances to pass through the walls thereof. The sachets each contain thermoplastic particles which have contained in the interstices thereof fragrance materials. The polymer particles can be foamed particles produced using chemical blowing agents or direct gas extrusion processes. Also described are methods for fabricating such sachets and methods of using same.</p>		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece			TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon			PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

- 1 -

**TIME RELEASE FRAGRANCE SACHET, METHOD OF USING SAME
AND METHOD OF FABRICATING SAME**

BACKGROUND OF THE INVENTION

5

This invention relates to time release fragrance sachets, methods of using same and methods of fabricating same. The sachets contain thermoplastic polymeric particles, including polymeric foam particles as control release compositions for use in the sachets.

10

An ever increasing requirement for air fresheners, including air fresheners for stored clothing exists for a slow controlled release device for slowly and controllably releasing fragrances into a gaseous environment in order to freshen air and in order to prolong the just washed fragrance/freshness of clothing and household linens.

15

Slow release polymers containing perfumes are well known in the prior art. Thus, United Kingdom Patent Specification No. 1,589,201 assigned to Hercules, Inc. discloses a thermoplastic resin body consisting of a thermoplastic polymer of ethylene and 6-60 weight percent of a polar vinyl monomer selected from the group consisting of vinyl acetate, methyl acrylate, ethyl acrylate, butyl acrylate and acrylic acid wherein the perfumed resin body is suitable for the preparation of shaped objects from which perfume odor emanates over a prolonged period at a stable level.

25

U.S. Patent No. 3,505,432 discloses a method of scenting a polyolefin which comprises:

30

- 2 -

- 5 (a) mixing a first amount of liquid polyolefin, e.g., polyethylene or polypropylene with a relatively large amount of scent-imparting material to form a flowable mass;
- (b) forming drops from said mass and causing substantially instantaneous solidification of said drops into polyolefin pellets having a relatively large amount of scent-imparting material imprisoned therein;
- 10 (c) melting said pellets with a second amount of said polyolefin, said second amount being larger than said first amount; and
- 15 (d) solidifying the melt of (c).

U.S. Patent No. 4,247,498 issued on January 27, 1981 discloses microporous polymers which are capable of containing volatile substances such as perfumes and the like in forms ranging from films to blocks in intricate shapes from synthetic thermoplastic polymers such as olefinic, condensation or oxidation polymers. In one embodiment of U.S. Patent No. 4,247,498, the microporous polymers are characterized by relatively homogeneous three-dimensional cellular structure having cells connected by pores of smaller dimension. Also disclosed in U.S. Patent No. 4,247,498 is a process for making microporous polymers from such thermoplastic polymers by heating a mixture of the polymer and compatible liquid (e.g., a perfume substance or the like) to form a homogeneous solution, cooling said

20

25

30

- 3 -

solution under non-equilibrium thermodynamic conditions to initiate liquid-liquid phase separation and continuing said cooling until the mixture achieves substantial handling strength. Also disclosed in said U.S. Patent No. 4,247,498
5 are microporous polymer products which contain relatively large amounts of such functionally useful fluids as perfume compositions and behave as solids.

U.S. Patent No. 4,156,067 issued on May 22, 1979 discloses
10 polyurethane polymers characterized by a molecular weight of above 6,000 and having lactone groups and hydroxyl groups in the polymer backbone being prepared by reacting a mixture of polyols, a polyfunctional lactone (e.g., epsilon caprolactone) and a polyfunctional isocyanate proportioned
15 so as to provide certain desired polymer properties. It is indicated in said U.S. Patent No. 4,156,067 that the product is soluble in alkaline solutions and may be used for light sensitive photographic layers on films, paper or glass; in drug delivery systems, as burn dressings; in body implants
20 such as vascular prosthesis; in molding compositions; and in the manufacture of catheters as well as in delivery of perfume compositions in a slow release manner. It is further indicated in said U.S. Patent No. 4,156,067 that the water absorptivity of the polyurethane/lactone polymers is
25 above 10%, preferably in the range of about 20% to 60%, and these polymers may range in their physical properties from rigid solids to completely gel-like high water absorptive polymers. It is further indicated in said U.S. Patent No. 4,156,067 that the polymers provide a leachable substrate
30 wherein the leaching agent may be water, gases, alcohols, esters and body fluids, e.g., animal or human.

- 4 -

Extrusion of thermoplastic foams is well known in the prior art. Thus, the *Modern Plastics Encyclopedia* (published by the McGraw-Hill Publishing Company), 1982-1983 edition at
5 pages 274 and 275 discloses a section authored by Fred Schrafft entitled "Extruding Thermoplastic Foams." Said article on pages 274 and 275 is incorporated by reference herein. It is indicated therein that three different processes are used for the extrusion of thermoplastic foams:

10

- (i) extrusion of expandable beads;
- (ii) extrusion of thermoplastics containing a chemical blowing agent; and
- (iii) direct gas extrusion process.

15

It is further indicated in the Schrafft article that the extrusion using a chemical blowing agent may be carried out on a normal single screw extruder, and the direct gas extrusion process may be carried out on single and twin
20 screw extruders. It is further indicated in the Schrafft article that common blowing agents used in the process are hydrocarbons such as pentene or fluorocarbons such as 11, 12 and 114. It is further stated that:

25

"the amount of blowing agent can vary widely depending on the resin and the type of product desired. However, generally about 7% blowing agent produces a product of about 5.6 lbs/cu. ft., while 18% blowing agent produces a product of about 1.9 lbs/cu. ft. ..."

30

- 5 -

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1A represents a perspective view of the outside of a foamed polymeric particle containing fragrance produced according to the process of Example I wherein a nitrogen foaming agent and a fragrance for use in sachets were added to an extruder during the extrusion of polyethylene.

Figure 1B is a cutaway side elevation view of the particle of Figure 1A.

Figure 2 is a cutaway side elevation schematic diagram of a screw extruder during the compounding of the resin with a fragrance while simultaneously adding foaming agent into the hollow portion of the barrel of the extruder and incorporates the pelletizing apparatus used in pelletizing the extruded foamed tow produced as a result of the extrusion operation.

Figure 3 is a cutaway perspective diagram of a pelletizing apparatus used in conjunction with the extrusion apparatus, for example, that illustrated in Figure 2, whereby the extruded tow is pelletized.

Figure 4 is a cutaway side elevation view of extrusion apparatus used for extruding thermoplastic polymeric foamed tubing containing within the walls of the tubing fragrance for use in the sachets of our invention.

Figures 5A and 5B represent cutaway side elevation views of injection molding apparatus prior to and during the

- 6 -

injection molding operation for the injection molding of fragrance-containing foamed polymeric pellets useful in practicing our invention.

5 Figure 5A shows the apparatus immediately prior to the carrying out of the injection molding process and Figure 5B shows the apparatus during the injection molding process wherein the polymeric foamed pellets are being fused and pushed through the injection molding apparatus orifice into
10 the mold.

Figure 6 is a cutaway side elevation view of injection molding apparatus useful in forming articles from the polymeric pellets containing fragrance useful in practicing
15 our invention.

Figure 7 is a cutaway side elevation schematic view of jet molding apparatus useful in forming articles of manufacture from the polymeric pellets containing fragrance.

20

Figure 8 is a partially cutaway perspective view of an article of manufacture useful as a sachet.

Figure 9 is a cutaway side elevation view of the article of manufacture of Figure 8 looking in the direction of the
25 arrows.

Figure 10 is a top view of the article of manufacture of Figure 8.

30

- 7 -

Figure 11 is a cutaway perspective view of a sachet of our invention showing in schematic form the controllable release of fragrance through the walls of the sachet product.

5 Figures 12A and 12B show prefabrication portions of the sachet of Figure 11. Figure 12A shows one part of the pre-formed sachet containing fragrance-containing polymeric particles ready to be heat sealed to the second part of the sachet to form the sachet of Figure 11. Figure 12B shows
10 the other part of the sachet prior to forming, ready to be heat sealed to that part of the sachet as illustrated in Figure 12A.

15 Figure 13 sets forth a diagram indicating the symbols used in the drawings of Figures 13A, 13B, 13C and 13D.

20 Figure 13A sets forth a comparison of in-use products of Figure 13, showing for each product strength on a scale of zero to 5 versus time. Figures 13B, 13C and 13D show graphs similar to those of Figure 13A with different concentrations of fragrance and different amounts of perfumed polymer located within each sachet.

25 Figure 14 is a diagram setting forth the scale used for Figures 14A, 14B, 14C, 14D and 14E.

30 Figures 14A, 14B, 14C, 14D and 14E set forth in-use graphs for sachets containing the products as set forth in Figure 14 with strength on a scale of zero to 5 on the Y axis and time being set forth on the X axis of each graph.

- 8 -

Figure 15 sets forth another scale used for the graphs of Figures 13A, 13B, 13C, 13D, 14A, 14B, 14C, 14D and 14E.

In summary, the graphs of Figure 13A, 13B, 13C and 13D show
5 the performance of mixtures of perfumed polymer and
vermiculite (expanded clay) with different perfume loadings
(10% and 20%) and different weights of particles (7 grams
and 14 grams). The graphs of Figures 14A, 14B, 14C and 14D
show sachet performance with and without liquid fabric
10 softener. The graph of Figure 14E shows lavender sachet
performance at different weights and perfume loadings.

Figure 16 is a partial side elevation view and partial
sectional view of an apparatus for forming non-foamed
15 polymer pellets containing fragrances useful in the practice
of our invention.

Figure 17 is a section taken along line 17-17 of Figure 16.

20

SUMMARY OF THE INVENTION

Our invention concerns time release fragrance sachets,
methods of using same and methods of fabricating same.

25

The time release sachets of our invention comprise hollow
container means fabricated from porous or non-porous
materials. The porous materials are porous to the
fragrances contained within perfumed polymer particles
30 located within the sachet.

- 9 -

The perfumed particles are fabricated using extrusion techniques and according to techniques as set forth in U.S. Letters Patent No. 4,521,541 issued on June 4, 1985, the specification for which is incorporated by reference herein;
5 U.S. Letters Patent No. 4,542,162 issued on September 17, 1985, the specification for which is incorporated by reference herein; as well as U.S. Letters Patent No. 5,543,398 issued on August 6, 1996, the specification for which is incorporated by reference herein.

10

The polymer particles, containing the perfumes located within the sachet, contain perfumes as defined in U.S. Letters Patent No. 5,540,853 issued on July 30, 1996, the specification for which is incorporated by reference herein.

15

In one embodiment, the perfume particles useful in the practice of our invention are "puffed" as a result of gaseous entrainment, which is carried out during the formation of said particles by means of extrusion.

20

Preferably, the sachets of our invention are fabricated from perfume-porous materials which typically have heat sealed closures. However, the enclosures of our invention may be fabricated from non-porous materials if the walls have
25 discrete openings so that fragrances may pass therethrough as they are being emitted from the control release fragrance-containing polymeric particles. Further, various forms of sealing the sachets are envisaged.

30

In a further embodiment of the invention, the sachet according to the invention may include a desiccant such as

- 10 -

for example silica gel. Further, the sachets may include an anti-moth agent, typically in the form of a perfume.

Examples of perfume-porous materials are synthetic
5 non-woven polyesters, synthetic non-woven polypropylene,
non-woven viscose and natural woven cotton interlock
materials. The sachet enclosures are fabricated from
substances having from about 10 to 300, preferably 30 up to
about 120 grams per square centimeter-(gsm) as a measurement
10 of the weave. The geometry of the sachets of our invention
may be spherical, ellipsoidal, cylindrical or conical. The
dimensions may be such that the height may vary from about 3
inches up to about 6 inches; the width may vary from about 2
inches up to about 5 inches; and, in the case of a spherical
15 sachet, the diameter may vary from about 3 inches up to
about 5 inches.

The perfumed polymer particle weight within each sachet may
vary from about 2 grams up to about 50 grams, with a
20 preferred range of from about 5 grams up to about 15 grams
of perfumed particle. The size of each perfumed particle
may vary from about 0.05 cm up to about 1 cm.

The fragrance material within the polymer has a calculated
25 $\log_{10}P$ in the range of from about 3 up to about 8, wherein P
is the partition coefficient of the active or bioactive
material between n-octanol and water.

The perfume ingredients are preferred to have a boiling
30 point $\geq 250^{\circ}\text{C}$. The $\log P$ of many perfume ingredients has
been reported; for example, the Pomona92 database, available

- 11 -

from Daylight Chemical Information Systems, Inc. (Daylight CIS), Irvine, California, contains many, along with citations to the original literature. However the logP values are most conveniently calculated by the "CLOGP" program, also available from Daylight CIS. This program also lists experimental logP values when they are available in the Pomona92 database. The "calculated logP" (ClogP) is determined by the fragment approach of Hansch and Leo (cf., A. Leo, in *Comprehensive Medicinal Chemistry*, Volume 4, C. Hansch, P.G. Sammens, J.B. Taylor and C.A. Ramsden, editors, page 295, Pergamon Press, 1990, incorporated by reference herein). The fragment approach is based on the chemical structure of each perfume ingredient and takes into account the numbers and types of atoms, the atom connectivity and the chemical bonding. The ClogP values, which are the most reliable and widely used estimates for this physiochemical property, are preferably used instead of the experimental logP values in the selection of perfume ingredients which are useful in the present invention.

Non-enduring perfume ingredients, which are preferably minimized in personal treatment, e.g., liquid personal cleansing compositions of the present invention, are those having a B.P. of less than about 250°C or having a ClogP of less than about 3.0 or having both a B.P. of less than about 250°C and a ClogP of less than about 3.0. The table below gives some non-limiting examples of non-enduring perfume ingredients. In some particular fabric softener compositions, some non-enduring perfume ingredients can be used in small amounts, e.g., to improve product odor. However, to minimize waste, the enduring perfume compositions of the

- 12 -

present invention contain less than about 30 weight percent of non-enduring perfume ingredients, preferably less than about 25 weight percent of non-enduring perfume ingredients, more preferably, less than about 20 weight percent of non-
5 enduring perfume ingredients, and even more preferably, less than about 15 weight percent of non-enduring perfume ingredients.

Examples of perfume materials which have a calculated logP \geq 3 are as set forth in the following table:

Examples of Enduring Perfume Ingredients		
Perfume Ingredients	Approximate B.P. ($^{\circ}$ C) ^(a)	ClogP
<u>B.P. > 250$^{\circ}$C and ClogP > 3.0</u>		
Allyl cyclohexane propionate	267	3.935
Ambrettolide	300	6.261
Amyl benzoate	262	3.417
Amyl cinnamate	310	3.771
Amyl cinnamic aldehyde	285	4.324
Amyl cinnamic aldehyde dimethyl acetal	300	4.033
iso-Amyl salicylate	277	4.601
Aurantiol	450	4.216
Benzophenone	306	3.120
Benzyl salicylate	300	4.383
para-tert-Butyl cyclohexyl acetate	+250	4.019
iso-Butyl quinoline	252	4.193
β -Caryophyllene	256	6.333
Cadinene	275	7.346
Cedrol	291	4.530
Cedryl acetate	303	5.436
Cedryl formate	+250	5.070
Cinnamyl cinnamate	370	5.480
Cyclohexyl salicylate	304	5.265
Cyclamen aldehyde	270	3.680
Dihydro isojasmonate	+300	3.009
Diphenyl methane	262	4.059
Diphenyl oxide	252	4.240
Dodecalactone	258	4.359
ISO E SUPER	+250	3.455
Ethylene brassylate	332	4.554
Ethyl methyl phenyl glycidate	260	3.165
Ethyl undecylenate	264	4.888
Exaltolide	280	5.346
GALAXOLIDE	+250	5.482
Geranyl anthranilate	312	4.216

- 14 -

Examples of Enduring Perfume Ingredients		
Perfume Ingredients	Approximate B.P. (°C) ^(a)	ClogP
<u>B.P. > 250°C and ClogP > 3.0</u>	+250	5.233
Hexadecanolide	294	6.805
Hexenyl salicylate	271	4.716
Hexyl cinnamic aldehyde	305	5.473
Hexyl salicylate	290	5.260
α -Irone	250	3.820
Lilial (p-t-bucinal)	258	3.858
Linalyl benzoate	263	5.233
2-Methoxy naphthalene	274	3.235
Methyl dihydrojasmone	+300	4.843
γ -n-Methyl ionone	252	4.309
Musk indanone	+250	5.458
Musk ketone	MP = 137°C	3.014
Musk tibetine	MP = 136°C	3.831
Myristicin	276	3.200
Oxahexadecanolide-10	+300	4.336
Oxahexadecanolide-11	MP = 35°C	4.336
Patchouli alcohol	285	4.530
Phantolide	288	5.977
Phenyl ethyl benzoate	300	4.058
Phenyl ethyl phenyl acetate	325	3.767
Phenyl heptanol	261	3.478
Phenyl hexanol	258	3.299
α -Santalol	301	3.800
Thibetolide	280	6.246
δ -Undecalactone	290	3.830

- 15 -

table - continued

Examples of Enduring Perfume Ingredients		
Perfume Ingredients	Approximate B.P. (°C) ^(a)	ClogP
<u>B.P. > 250°C and ClogP > 3.0</u>		
γ -Undecalactone	297	4.140
Vetiveryl acetate	285	4.882
Yara-yara	274	30235
Ylangene	250	6.268

(a) M.P. is melting point; these ingredients have a B.P.
higher than 250°C.

5

The amount of fragrance material in the perfumed polymer particles may vary from about 5% up to about 45% by weight of the perfumed particle.

10 As set forth, *supra*, foamed or non-foamed polymeric particles may be utilized in the practice of our invention.

The advantages of using the foamed polymeric particles are multiple, to wit:

15

- (a) improved handling;
 - (b) greater retention of fragrance when not in use;
and
 - (c) greater length of time during which release of
- 20 fragrance from polymer is at "steady state" or
"zero order."

- 16 -

Whether producing foamed or non-foamed polymer, the nature of the extruder utilized in the process for producing polymeric particles of our invention may be either single screw or double screw. Thus, the types of extruders that
5 can be used are disclosed at pages 246-267 and 332-349 of the *Modern Plastics Encyclopedia*, 1982-1983 (published by the McGraw-Hill Publishing Company), the disclosure of which is incorporated by reference herein. More specifically, examples of extruders which are usable in carrying out the
10 process for producing the polymeric particles used in our invention are as follows:

1. The Welex "Super Twinch" 3.5 inch extruder manufactured by Welex Incorporated, 850 Jolly Road, Blue Bell,
15 Pennsylvania 19422;
2. The Krauss-Maffei twin screw extruder manufactured by the Krauss-Maffei Corporation/Extruder Division, 3629 West 30th Street South, Wichita, Kansas 67277;
20
3. Modified Sterling model 4000 and 5000 series extruder manufactured by Sterling Extruder Corporation of 901 Durham Avenue, South Plainfield, New Jersey;
- 25 4. The CRT ("Counter-Rotating Tangential") Twin Screw Extruder manufactured by Welding Engineers, Inc., King of Prussia, Pennsylvania 19406;
- 30 5. The Leistritz Twin Screw Dispersion Compounder manufactured by the American Leistritz Extruder Corporation, 198 U.S. Route 206 South, Sommerville, New Jersey 08876;

- 17 -

6. The ZSK Twin Screw Co-Rotating Extruder manufactured by the Werner & Pfleiderer Corporation, 663 East Crescent Avenue, Ramsey, New Jersey 07446;

5

7. The Farrel Extruder manufactured by Farrel Connecticut Division, Emhart Machinery Group, Ansonia, Connecticut 06401;

10 8. The MPC/V Baker Perkins Twin Screw Extruder manufactured by the Baker Perkins Inc. Chemical Machinery Division, Saginaw, Michigan 48601; and

9. The Berstorff single screw, twin screw or foam
15 extrusion equipment manufactured by Berstorff Corporation, P.O. Box 240357, 8200-A Arrowridge Boulevard, Charlotte, North Carolina 28224; and

In producing the polymeric particles used in the sachets of
20 our invention, various polymers may be utilized, for example: low-density polyethylene, high-density polyethylene, polypropylene, the copolymer of ethylene and vinyl acetate and polyvinyl chloride. More specifically, the polymers used in the practice of our invention may be copolymers of
25 ethylene and a polar vinyl monomer selected from:

- (a) vinyl acetate;
- (b) ethyl acrylate;
- (c) methyl acrylate;
- 30 (d) butyl acrylate; and
- (e) acrylic acid,

- 18 -

including the hydrolyzed copolymer of ethylene and vinyl acetate. Preferred copolymers are ethylene vinyl acetate with about 9 to 60% vinyl acetate and ethylene/ethyl acrylate with about 6 to 18% ethyl acrylate.

Resins of the type disclosed for use as copolymers are commercially available in the molding powder form. For example, ethylene vinyl acetate copolymers are marketed by the E.I. duPont de Nemours Company under the trade name "ELVAX[®]" and by the Arco Polymer Division under the trademark "DYLAND[®]" and by the Exxon Corporation of Linden, New Jersey under the trademark "DEXXON[®]." Ethylene/ethyl acrylate copolymers are marketed by Union Carbide Corporation under the trade name "EEA RESINS[®]."

The polymer is added to the single screw or twin screw extruder at a feed rate in the range of from about 80 up to about 300 lbs/hour while maintaining the temperature in the screw extruder between about 160°C and about 240°C. If the polymer or copolymer powder is added to the extruder at a reference "barrel segment," then the fragrance is added to the extruder under pressure downstream from the addition point of the polymer at 1 or more of "barrel segments" 2-9.

The fragrance added at "barrel segments" 2-9 of the single screw or twin screw extruder then has one or more of the foregoing functions. Furthermore, the fragrance added at "barrel segments" 2-9 must be previously or made to be

- 19 -

compatible with the polymer added at "barrel segment" 1 of the single screw or twin extruder.

As stated, supra, various polymers are useful in the practice of our invention. Specific examples of polymers useful in the practice of our invention are as follows:

- 10 (a) DYLAN[®] brand of low density polyethylene. DYLAN is a trademark owned by the Atlantic Richfield Company of Los Angeles, California;
- (b) DYDLITE[®] brand of expandable polystyrene compositions. DYDLITE[®] is a trademark of the Atlantic Richfield Company of Los Angeles, California;
- 15 (c) SUPER DYLAN[®] brand of high density polyethylene. SUPER DYLAN[®] is a trademark of the Atlantic Richfield Company of Los Angeles, California;
- 20 (d) Blended polyethylene and carbon black as specifically taught in U.S. Patent No. 4,369,267 issued on January 18, 1983, the specification for which is incorporated by reference herein;
- (e) Polystyrene as disclosed in U.S. Patent No. 4,369,227 issued on January 18, 1983, the specification for which is incorporated by reference herein;
- 25 (f) Polyene/ α -olefin copolymers as exemplified and disclosed in U.S. Patent No. 4,369,291, the specification for which is incorporated by reference herein;

- 20 -

- (g) Poly- α -olefins disclosed in Canadian Patent No. 1,137,067 issued on December 7, 1982, the specification for which is incorporated by reference herein;
- 5 (h) Polymeric compositions as disclosed in Canadian Patent No. 1,137,068 issued on December 7, 1982, the specification for which is incorporated by reference herein;
- 10 (i) Poly- α -olefins disclosed in Canadian Patent No. 1,137,067, the specification for which is incorporated by reference herein;
- (j) Polyolefins described in Canadian Patent No. 1,137,066, the specification for which is incorporated by reference herein;
- 15 (k) Polyethylene oxides as disclosed in Canadian Patent No. 1,137,065 issued on December 7, 1982, the specification for which is incorporated by reference herein;
- 20 (l) Olefin polymers and copolymers as disclosed in Canadian Patent No. 1,139,737 issued on January 18, 1983, the disclosure of which is incorporated by reference herein.
- 25 (m) Polyolefins disclosed in Canadian Patent No. 1,139,738 issued on January 18, 1983, the specification for which is incorporated by reference herein.
- (n) Chlorinated PVC as disclosed in Polymer 1982.23 (7, Suppl.), 1051-6 abstracted at *CHEMICAL ABSTRACTS*, Volume 97:145570y, 1982;

- 21 -

- 5 (o) Polyepsilon caprolactone copolymers made by means of alcohol initiated polymerization as disclosed in *J. Polym. Sci., Polym. Chem. Ed.* 1982, 20(2), pages 319-26, abstracted at *CHEMICAL ABSTRACTS*, Volume 96:123625x, 1982;
- (p) Styrene acrylonitrile copolymers as disclosed in *Diss. Abstracts, Int. B*, 1982, 42(8), 3346 and abstracted at *CHEMICAL ABSTRACTS*, Volume 96:143750n, 1982;
- 10 (q) Copolymers of epsilon caprolactone with 1,4-butane diol as disclosed at *Kauch. Rezine*, 1982, (2), 8-9, abstracted at *CHEMICAL ABSTRACTS*, Volume 96:143750n, 1982;
- 15 (r) Polyesters as disclosed in U.S. Patent No. 4,326,010, the specification for which is incorporated by reference herein;
- (s) Chlorinated polyethylene as disclosed by Belorgey, et al, *J. Polym. Sci., Polym. Phys. Ed.* 1982, 20(2), 191-203
- 20 (t) Plasticized polyepsilon caprolactone copolymers containing dimethyl phthalate plasticizers as set forth in Japanese Patent No. J81/147844, abstracted at *CHEMICAL ABSTRACTS*, Volume 96:69984y (1982), the specification for which is
- 25 incorporated by reference herein;
- (u) Maleic anhydride modified adducts of polyepsilon caprolactone polyols and ethylenically unsaturated monomer as disclosed in U.S. Patent No. 4,137,279

- 22 -

issued on January 30, 1979, the specification for which is incorporated by reference herein;

5 (v) Polyurethane polymers having lactone backbones as disclosed in U.S. Patent No. 4,156,067 issued on May 22, 1979, the disclosure of which is incorporated by reference herein;

10 (w) Polyurethane polyether resins wherein the resin is obtained by reacting a polyfunctional lactone with a long chain polyalkylene diol and a urethane precursor as disclosed in U.S. Patent No. 4,355,550 issued on March 10, 1981, the disclosure of which is incorporated by reference herein; and

15 (x) Resins having polyurethane backbones as disclosed in U.S. Patent No. 3,975,350 issued on August 17, 1976, the disclosure of which is incorporated by reference herein.

DETAILED DESCRIPTION OF THE DRAWINGS

20 Figure 1A is an outer view of a foamed polymeric particle containing functional fluid or solid as indicated by reference numeral 10.

25 Figure 1B is a cross-section of the particle of Figure 1A, taken along line 1B in Figure 1A. Part of the particle indicated by reference numeral 10 is the outer surface thereof. Reference numeral 11 indicated one of the pores produced as a result of foaming.

- 23 -

CLAIMS

1. An article which is a fragrance sachet comprising:

- 5 (i) containment means consisting of a container wall separating an inner void from an outer environment; and
- (ii) contained within said inner void, a plurality
10 of extruded polymer fragrance particles

said container wall being capable of transmitting gaseous fragrance from said inner void to said outer environment and said perfumed polymer particles having a diameter of from
15 about 3 up to about 7 mm; said perfumed polymer particles containing from about 5% up to about 45% by weight of fragrance; and the fragrance contained in said perfumed particles having a calculated $\log_{10}P$ of from about 3 up to about 8 and a boiling point at atmospheric pressure of
20 greater than 250°C.

2. A three-dimensional sachet article located in a three-dimensional space comprising:

- 25 (i) hollow containment means consisting of a continuous thin lamina porous to perfumes having a calculated $\log_{10}P$ from about 3 up to about 8 substantially totally surrounding a specific internal void volume; and

30

- 24 -

- (ii) contained within, typically a major proportion of, said specific void volume and supported by said containment means a plurality of extruded polymeric particles, each of which contains within the interstices thereof from about 5 up to about 45% by weight of a perfume composition having a $\log_{10}P$ of from 3 up to about 8, which perfume composition is emitted in a controllably releasable manner over an extended period of time into said internal void volume and then through said containment means into said three-dimensional space.
- 15 3. A process for fragrancng a three-dimensional space over an extended period of time, comprising the step of placing the sachet article of Claim 1 within said three-dimensional space.
- 20 4. A process for fragrancng a three-dimensional space over an extended period of time, comprising the step of placing the sachet article of Claim 2 within said three-dimensional space.
- 25 5. A process for forming the article of Claim 2, comprising the steps of:
- (i) providing first planar lamina consisting of a polymer porous to perfumes having a calculated $\log_{10}P$ of from about 3 up to about 8 and having a circumference which is

- 25 -

sealable, ideally heat sealable, over
substantially its entire distance,;

- 5 (ii) placing on said first planar lamina a
plurality of extruded polymeric particles,
each of which contains within the interstices
thereof from about 5 up to about 45% by
weight of a perfume composition having a
calculated $\log_{10}P$ of from about 3 up to about
10 8, which particles emit said perfume
composition in a controllably releasable
manner over an extended period of time;
- 15 (iii) providing a second planar lamina having a
geometry substantially identical to said
first planar lamina, consisting of a polymer
porous to perfumes having a calculated $\log_{10}P$
of from about 3 up to about 8 and having a
circumference which is heat sealable
20 substantially over its entire length;
- (iv) placing said second planar lamina in contact
with said first planar lamina so that their
respective circumferences are juxtaposed with
25 one another; and
- (v) applying sufficient heat to an area proximate
said entire circumferences, whereby said
first planar lamina is sealed to said second
30 planar lamina along the entire circumferences

- 26 -

of said first planar lamina and said second planar lamina.

6. An article which is a fragrance sachet comprising:

5

(i) containment means consisting of a container wall separating an inner void from an outer environment; and

10

(ii) contained within said inner void, a plurality of extruded and puffed polymer fragrance particles

said container wall being capable of transmitting gaseous
15 fragrance from said inner void to said outer environment and
said perfumed polymer particles having a diameter of from
about 3 up to about 7 mm; said perfumed polymer particles
containing from about 5% up to about 45% by weight of
fragrance; and the fragrance contained in said perfumed
20 particles having a calculated $\log_{10}P$ of from about 3 up to
about 8 and a boiling point at atmospheric pressure of
greater than 250°C.

7. A three-dimensional sachet article located in a three-
25 dimensional space comprising:

(i) hollow containment means consisting of a
continuous thin lamina porous to perfumes
having a calculated $\log_{10}P$ from about 3 up to
30 about 8 substantially totally surrounding a
specific internal void volume; and

- 27 -

(ii) contained within a major proportion of said specific void volume and supported by said containment means a plurality of extruded and puffed polymeric particles, each of which contains within the interstices thereof from about 5 up to about 45% by weight of a perfume composition having a $\log_{10}P$ of from 3 up to about 8, which perfume composition is emitted in a controllably releasable manner over an extended period of time into said internal void volume and then through said containment means into said three-dimensional space.

8. A process for fragrancng a three-dimensional space over an extended period of time, comprising the step of placing the sachet article of Claim 6 within said three-dimensional space.

9. A process for fragrancng a three-dimensional space over an extended period of time, comprising the step of placing the sachet article of Claim 7 within said three-dimensional space.

10. A process for forming the article of Claim 2, comprising the steps of:

(i) providing first planar lamina consisting of a polymer porous to perfumes having a calculated $\log_{10}P$ of from about 3 up to about 8 and having a circumference which is heat

- 28 -

sealable over substantially its entire distance;

- 5 (ii) placing on said first planar lamina a plurality of extruded and puffed polymeric particles, each of which contains within the interstices thereof from about 5 up to about 45% by weight of a perfume composition having a calculated $\log_{10}P$ of from about 3 up to about 8, which particles emit said perfume composition in a controllably releasable manner over an extended period of time;
- 10
- 15 (iii) providing a second planar lamina having a geometry substantially identical to said first planar lamina, consisting of a polymer porous to perfumes having a calculated $\log_{10}P$ of from about 3 up to about 8 and having a circumference which is heat sealable substantially over its entire length;
- 20
- 25 (iv) placing said second planar lamina in contact with said first planar lamina so that their respective circumferences are juxtaposed with one another; and
- 30 (v) applying sufficient heat to an area proximate said entire circumferences, whereby said first planar lamina is sealed to said second planar lamina along the entire circumferences

- 29 -

of said first planar lamina and said second
planar lamina.

FIG. 1A

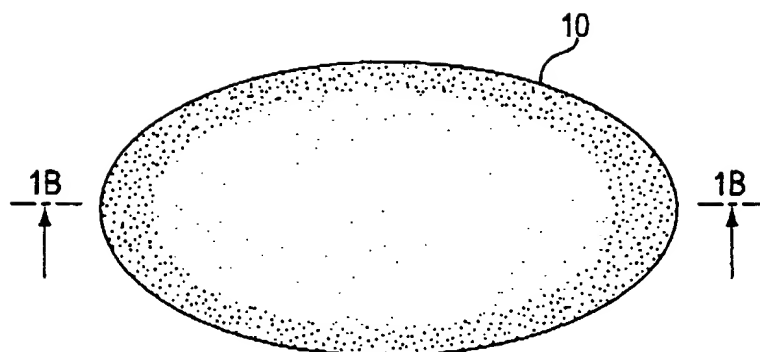


FIG. 1B

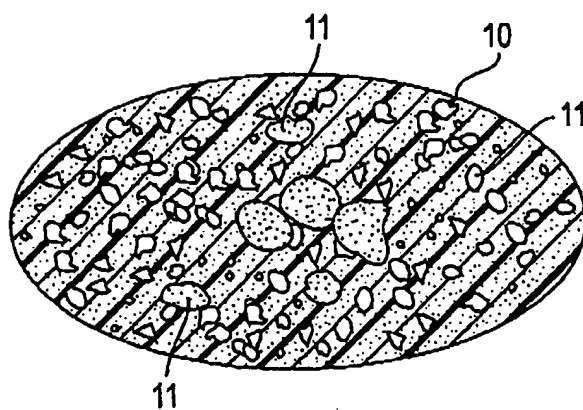


FIG. 2

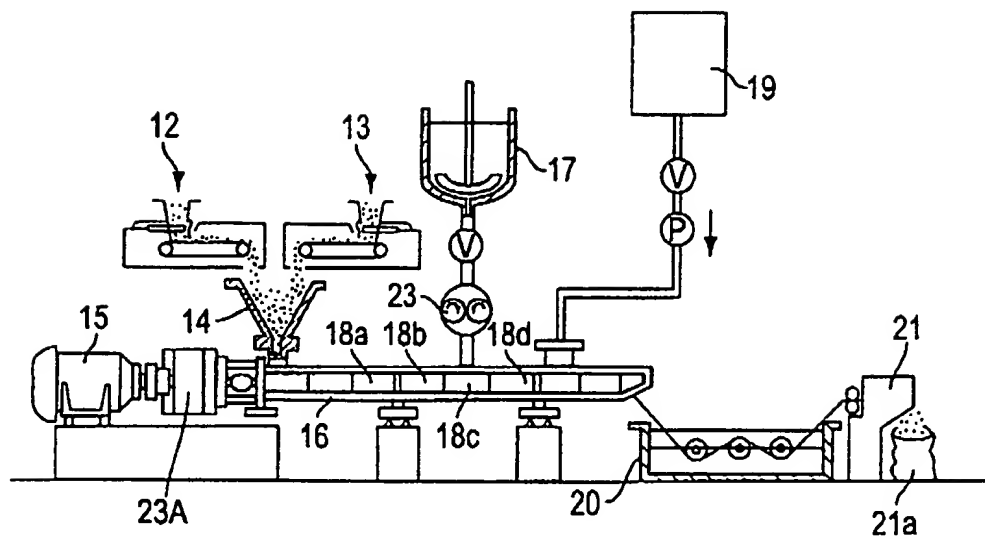


FIG. 3

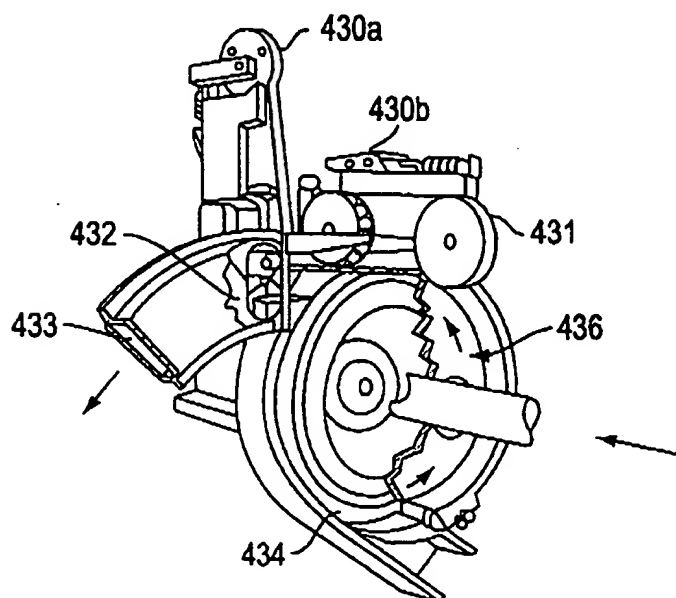


FIG. 4

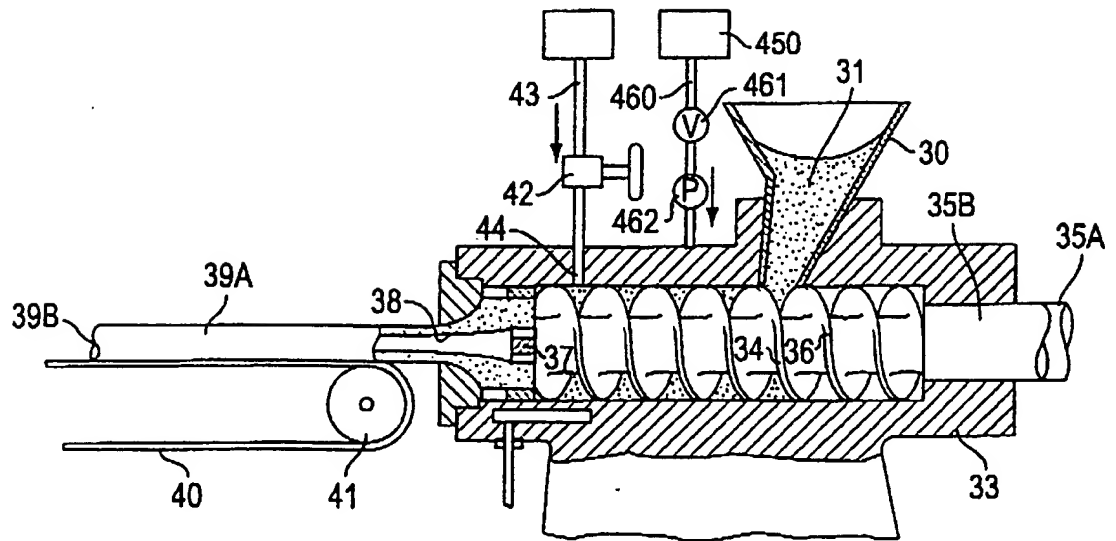


FIG. 5A

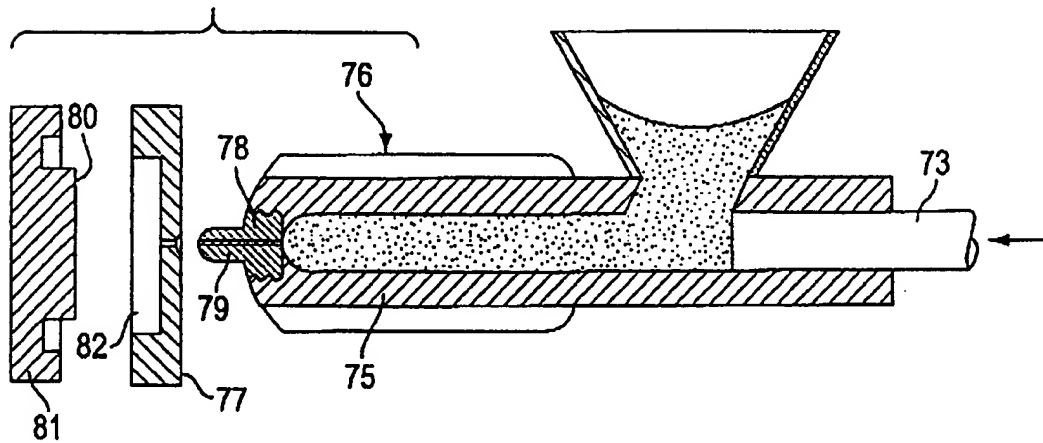


FIG. 5B

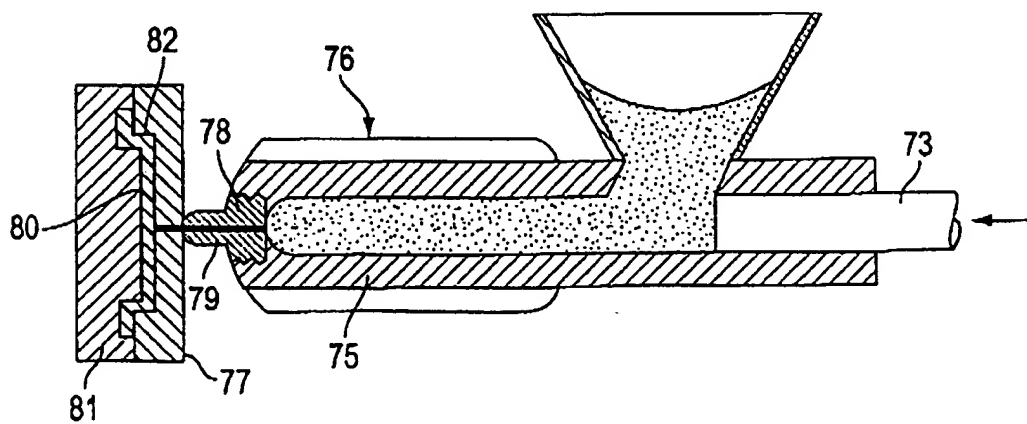


FIG. 6

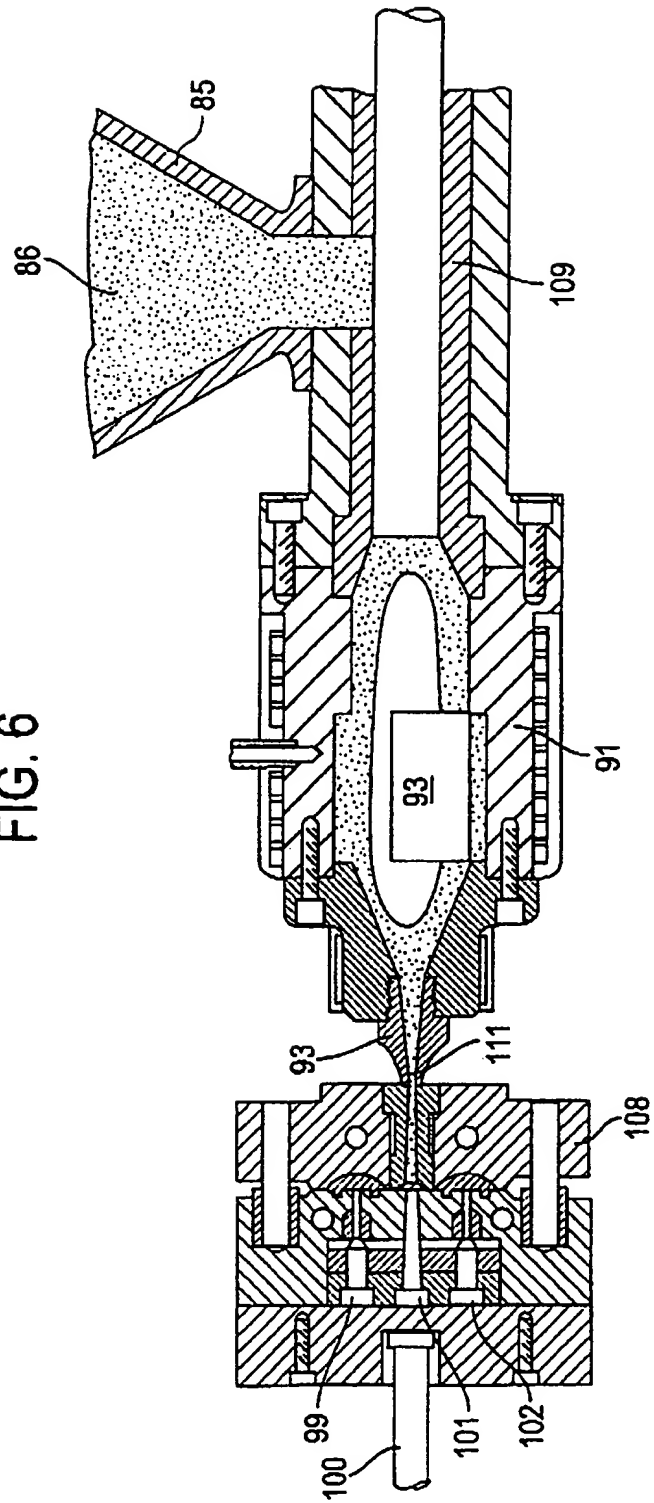


FIG. 7

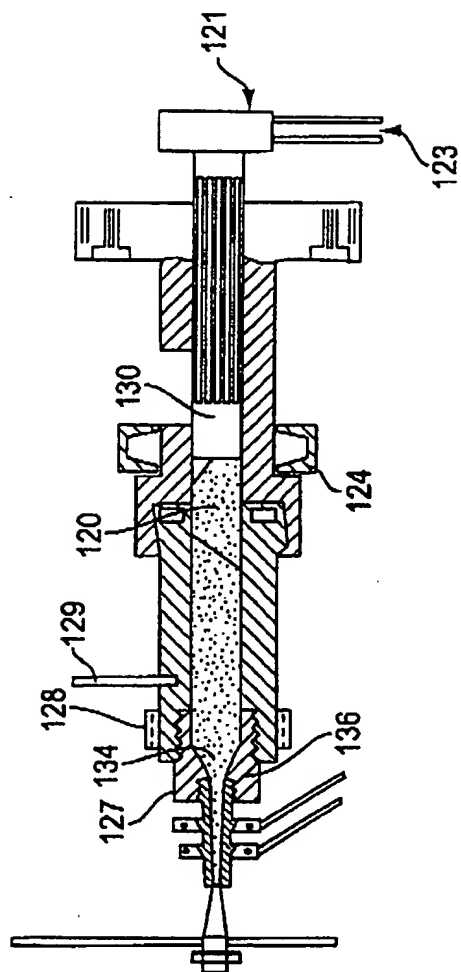


FIG.8

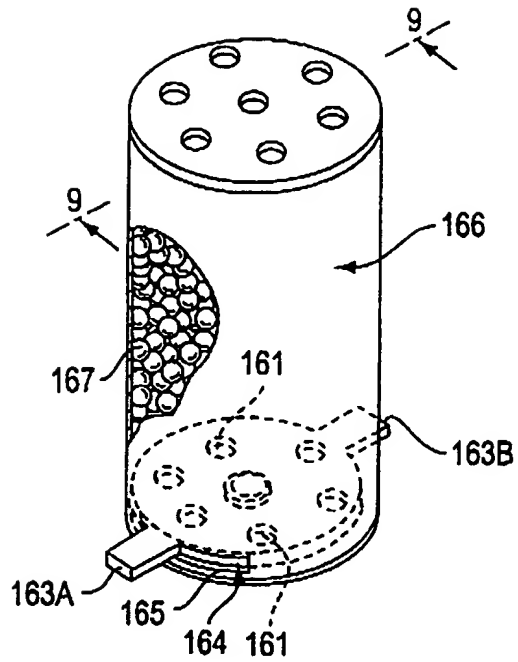


FIG.9

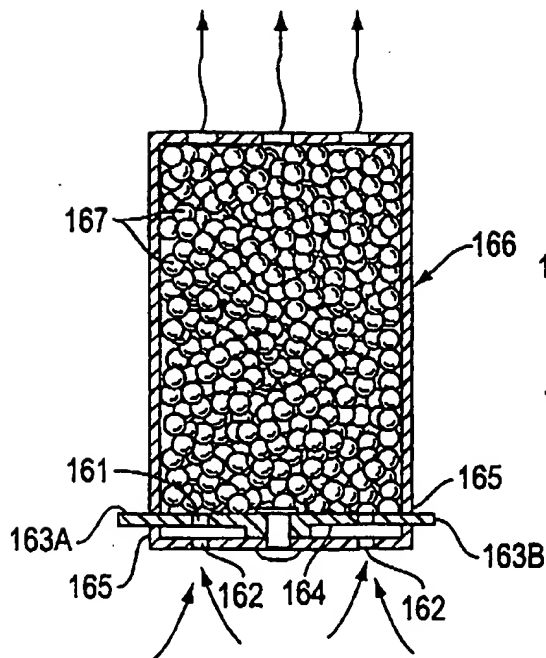


FIG.10

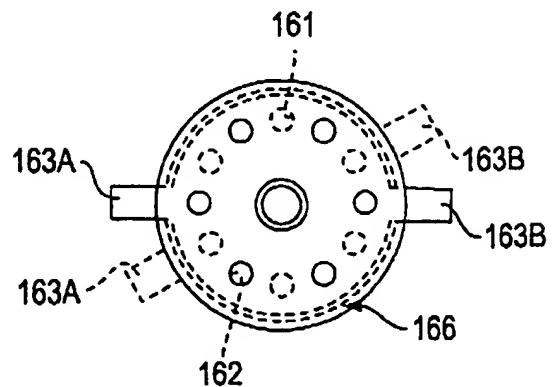


FIG. 11

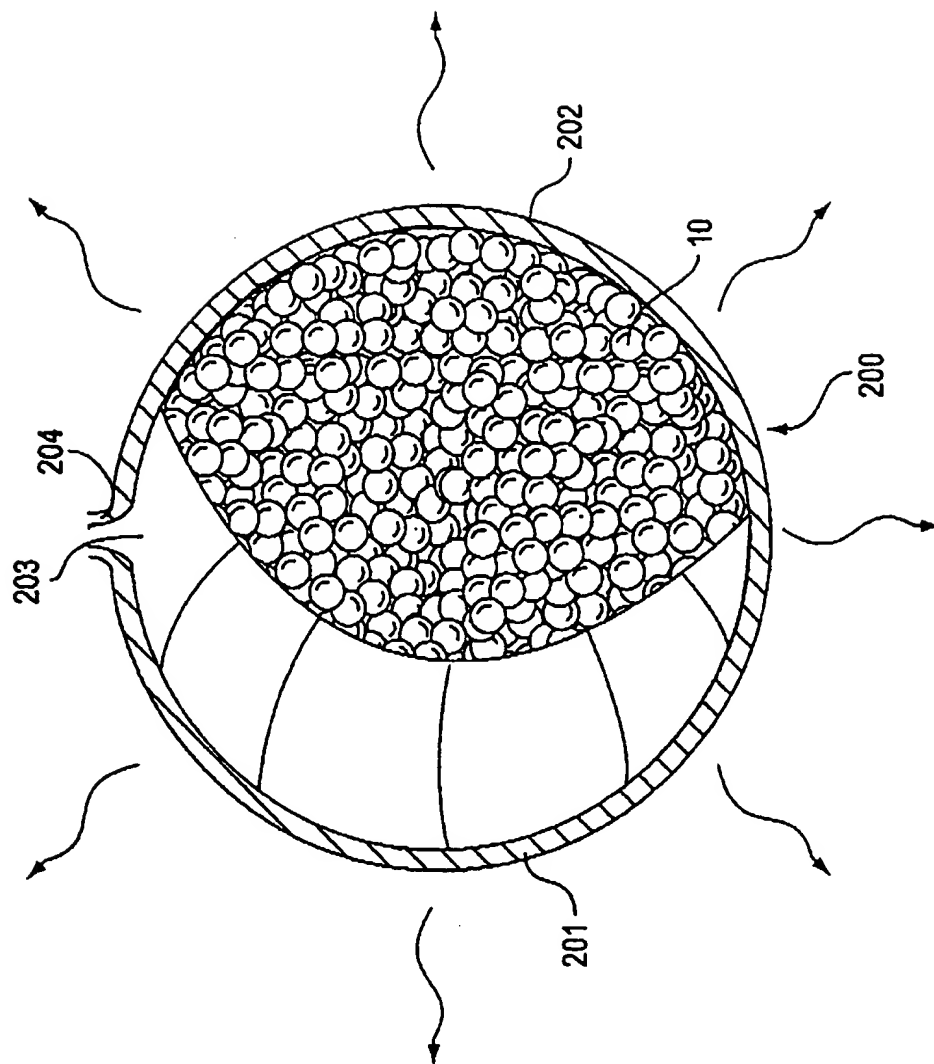


FIG. 12B

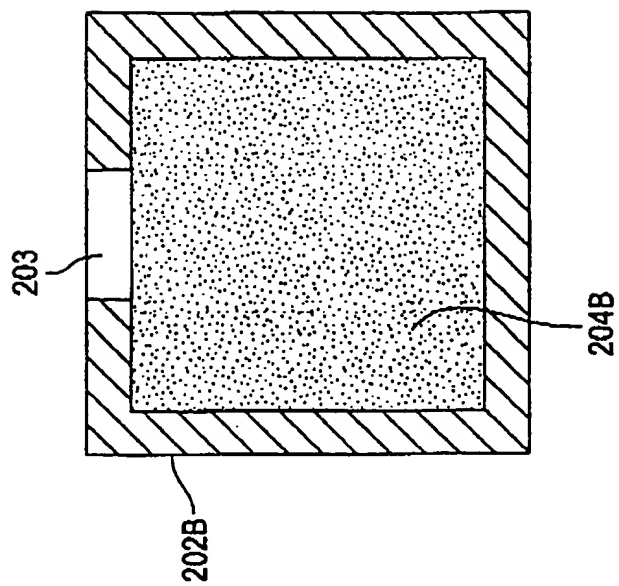


FIG. 12A

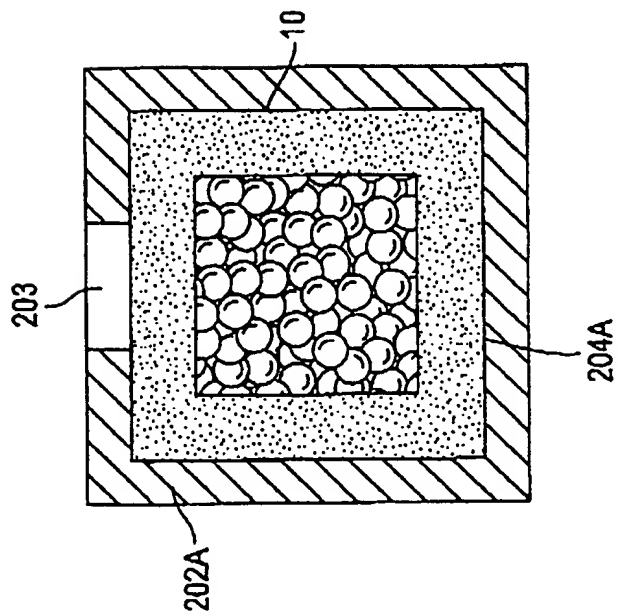


FIG. 13

7g V10	=100% PERFUMED VERMICULITE (10% PERFUME)
3g V10 + 3.5g P10	= MIX: 50% PERFUMED VERMICULITE (10% PERFUME AND 50% POLYIFF (10% PERFUME)
3.5g VO + 3.5g P20	=MIX: 50% UNPERFUMED VERMICULITE AND 50% POLYIFF (20% PERFUME)
7g P10	=100% POLYIFF (10% PERFUME)

FIG. 13A

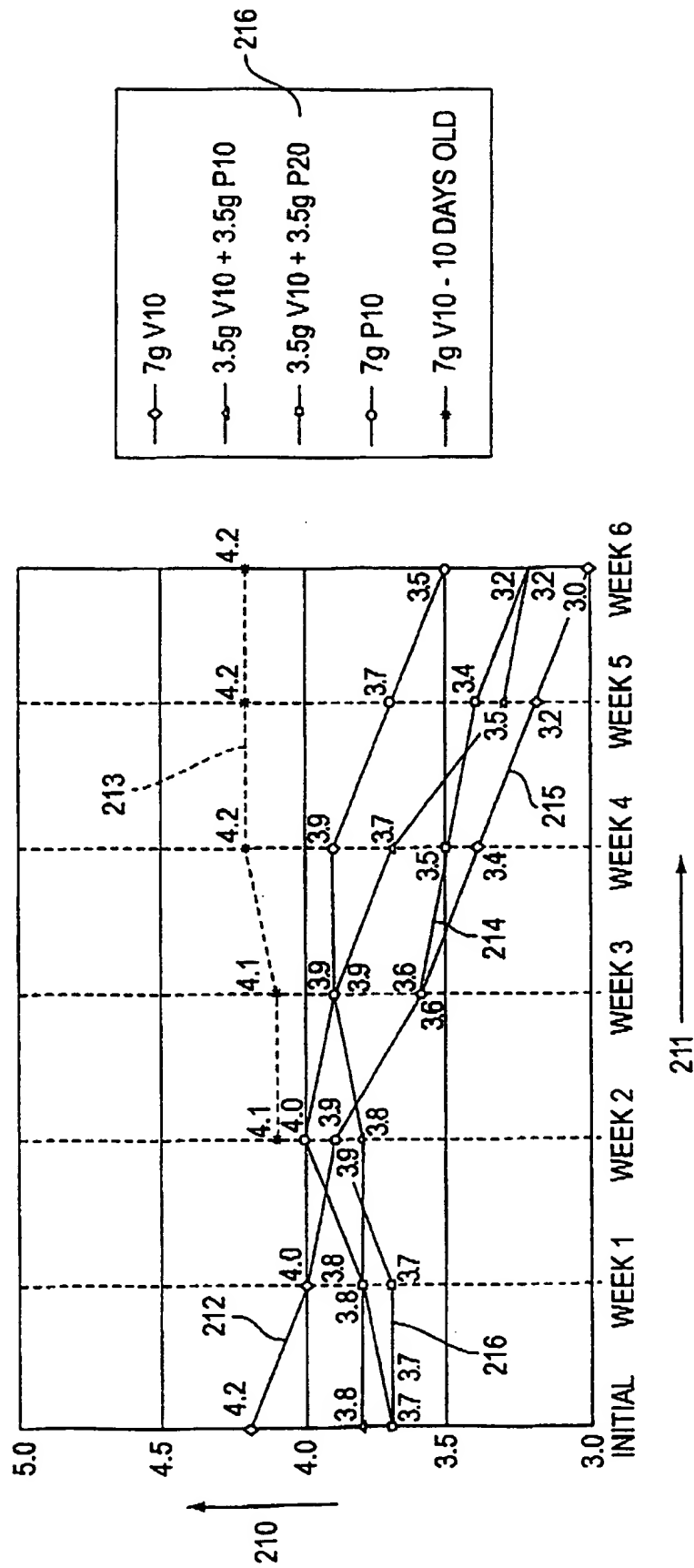


FIG. 13B

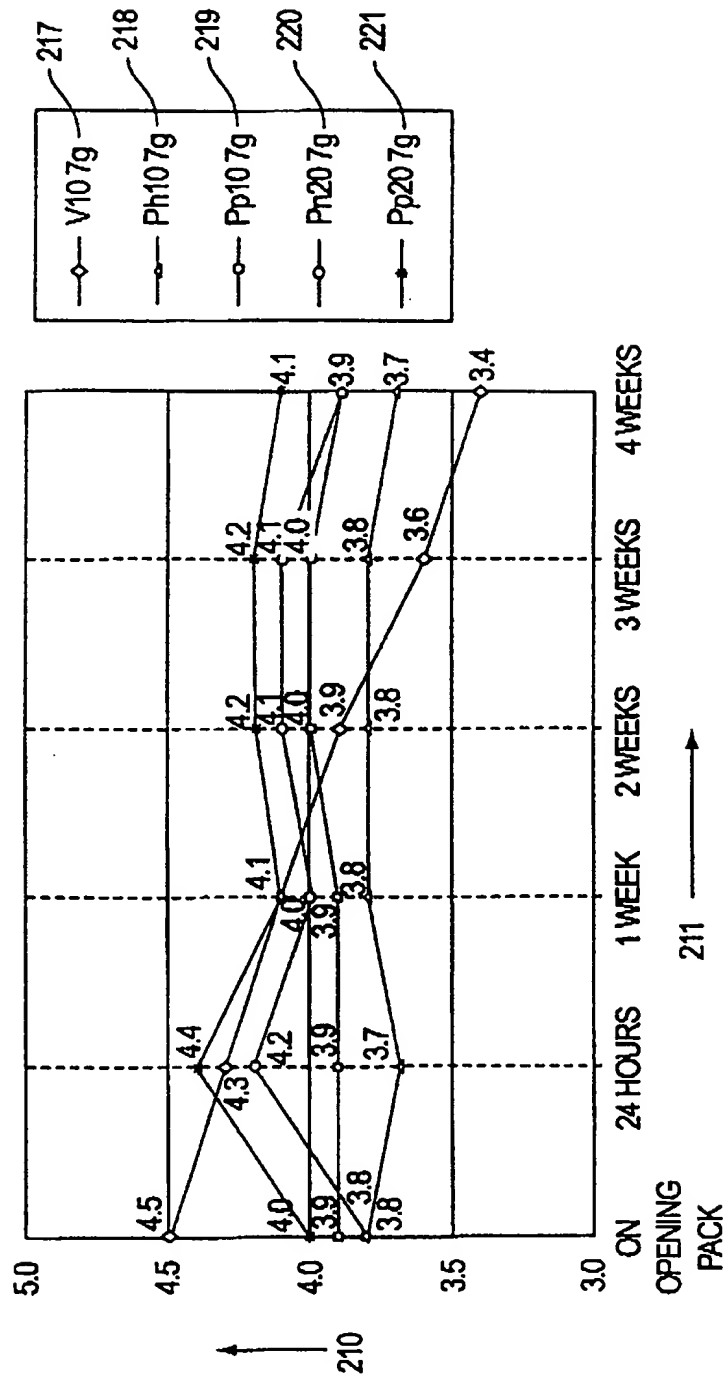


FIG 13C

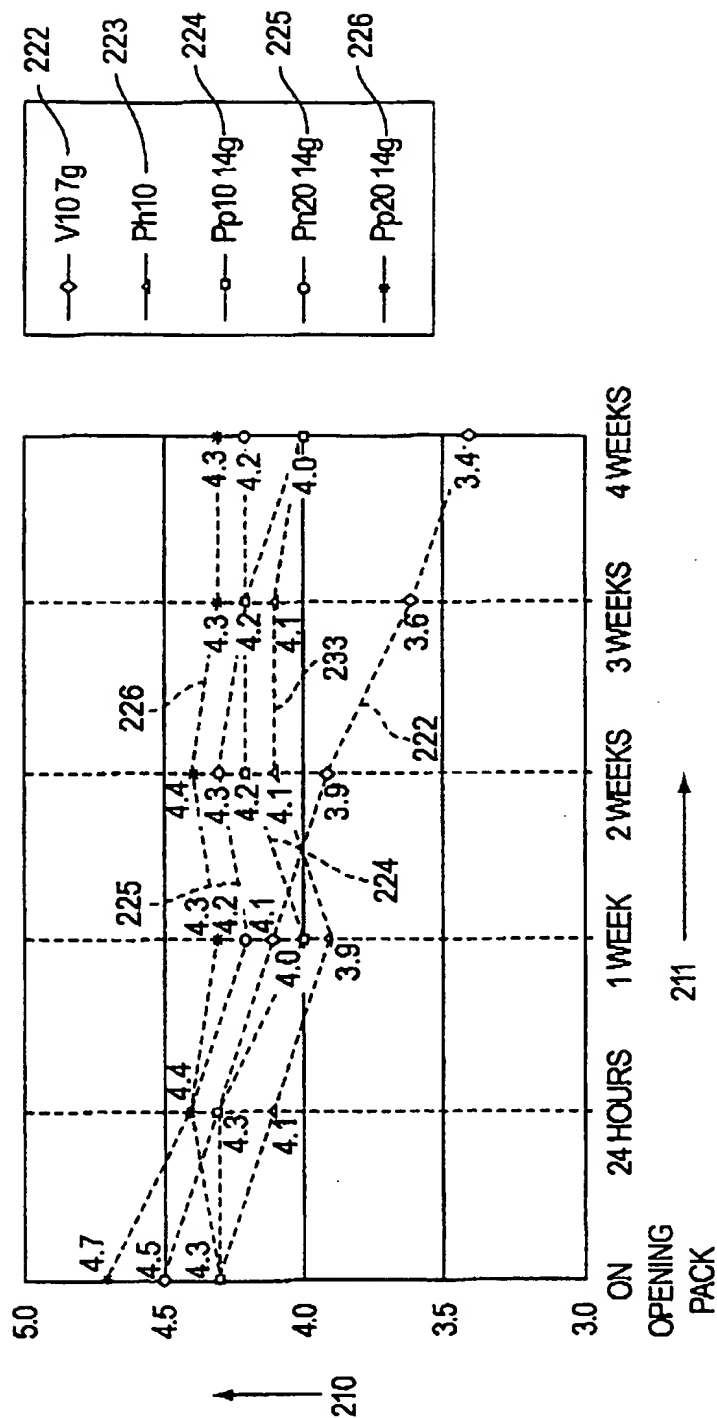


FIG. 13D

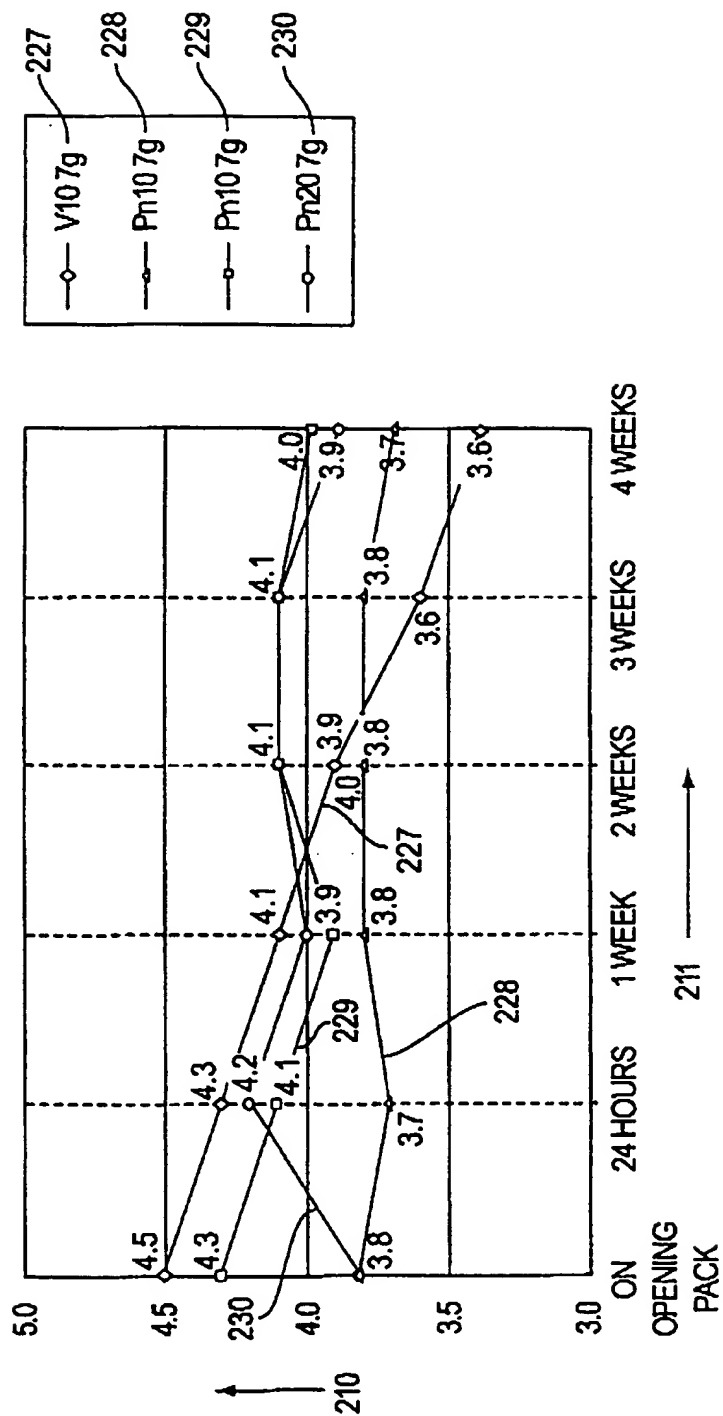


FIG. 14

0 = NO PERFUME

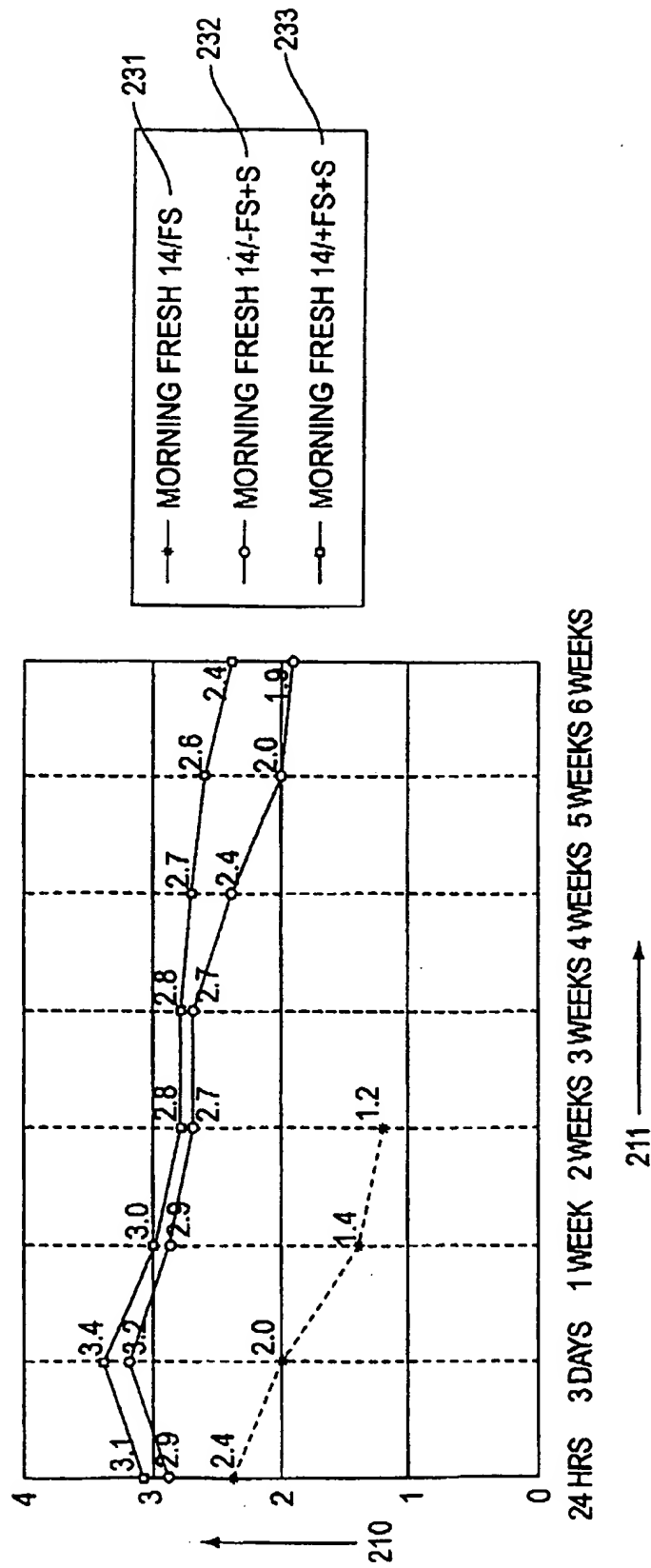
1 = SLIGHT PERFUME

2 = MODERATE PERFUME

3 = STRONG PERFUME

4 = VERY STRONG PERFUME

FIG. 14A

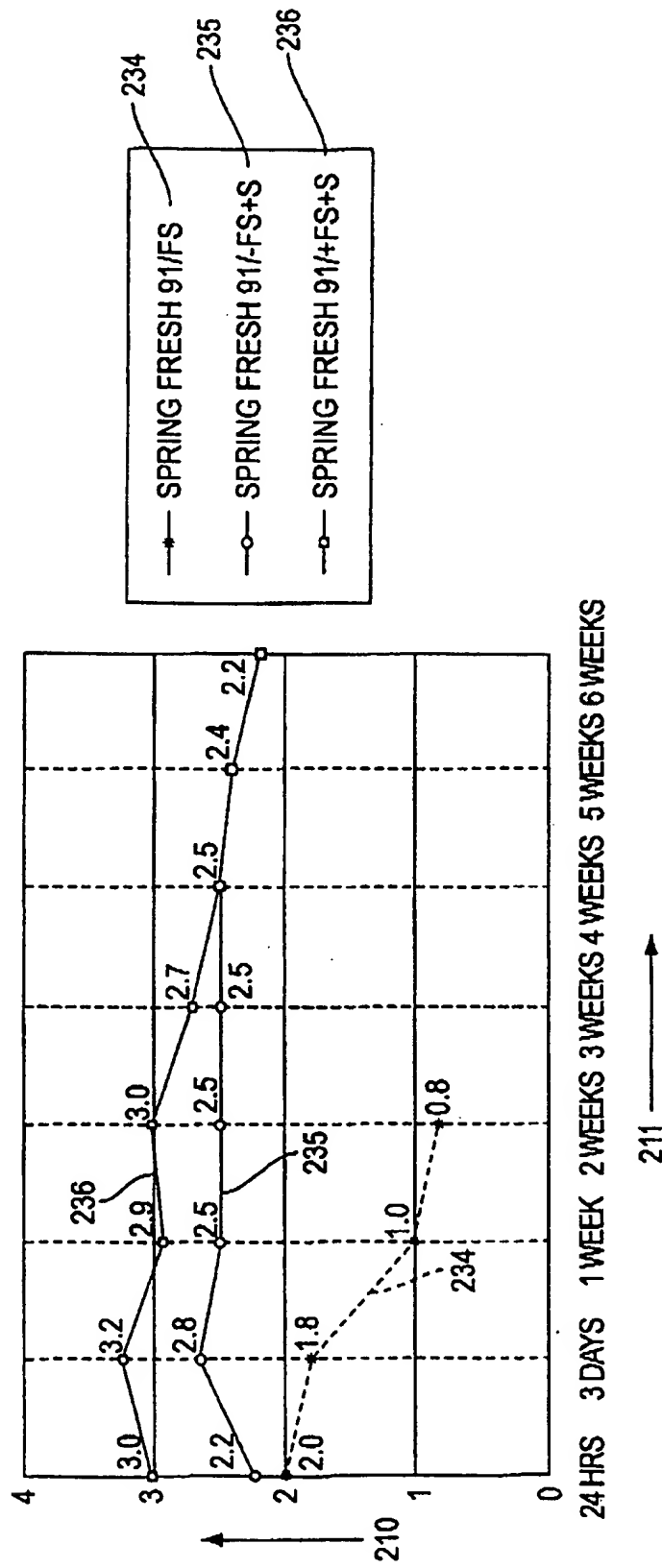


FS = FABRIC SOFTENER ONLY

-FS+S = NO FABRIC SOFTENER, SACHET ONLY

+FS+S = FABRIC SOFTENER AND SACHET

FIG. 14B

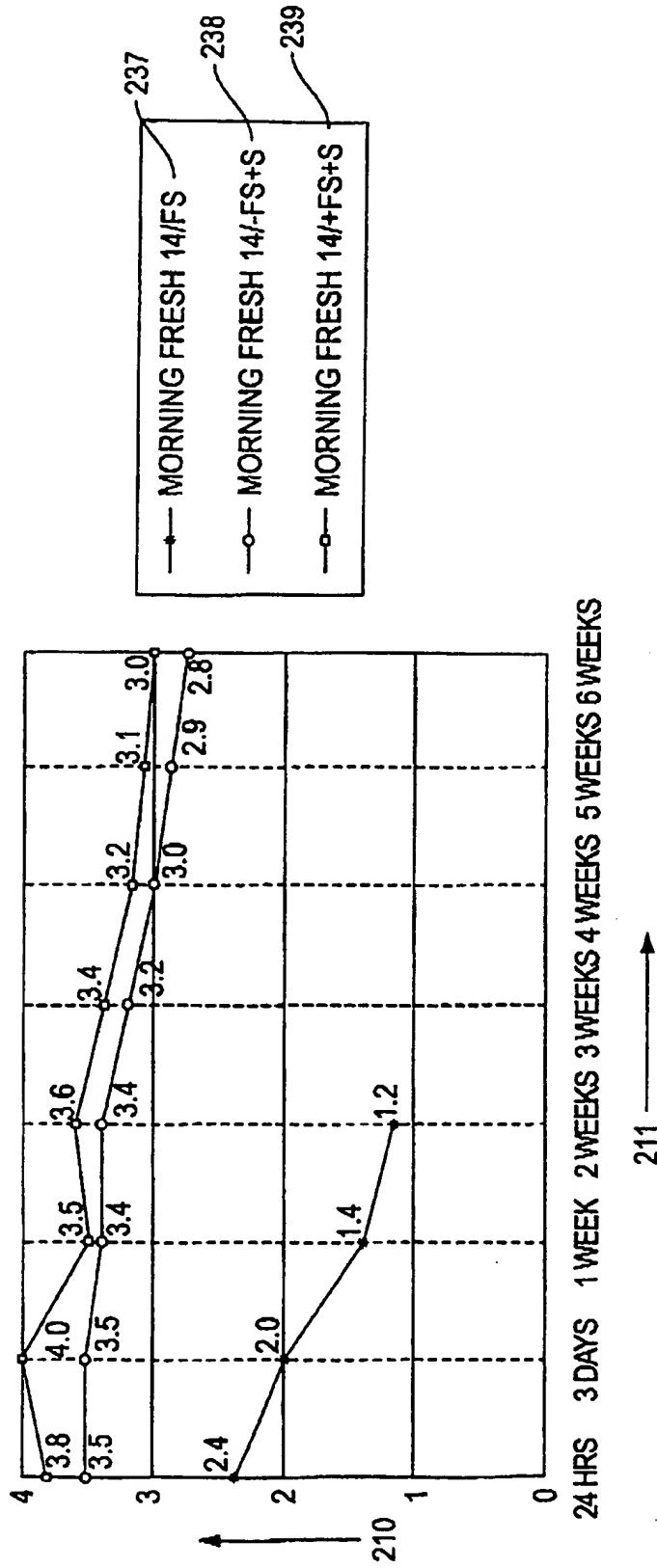


FS = FABRIC SOFTENER ONLY

-FS+S = NO FABRIC SOFTENER, SACHET ONLY

+FS+S = FABRIC SOFTENER AND SACHET

FIG. 14C

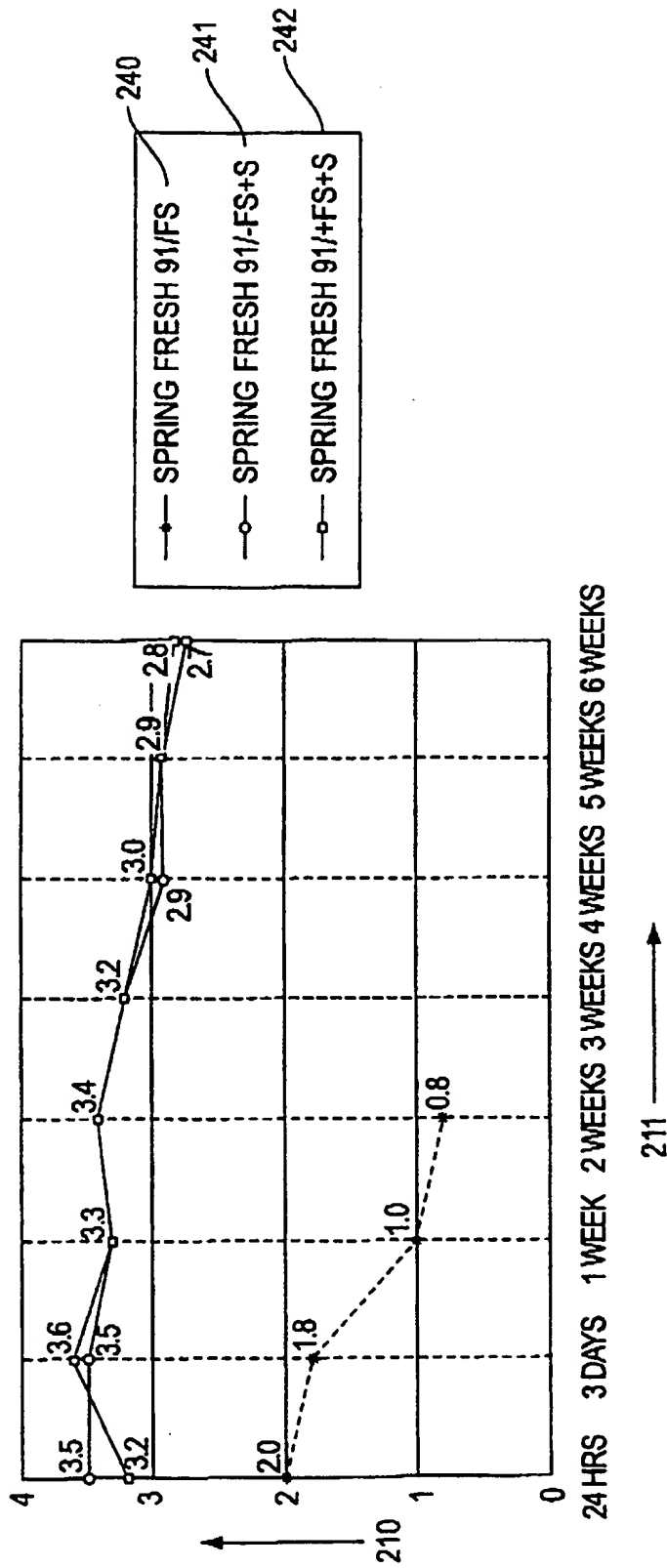


FS = FABRIC SOFTENER ONLY

-FS+S = NO FABRIC SOFTENER, SACHET ONLY

+FS+S = FABRIC SOFTENER AND SACHET

FIG. 14D

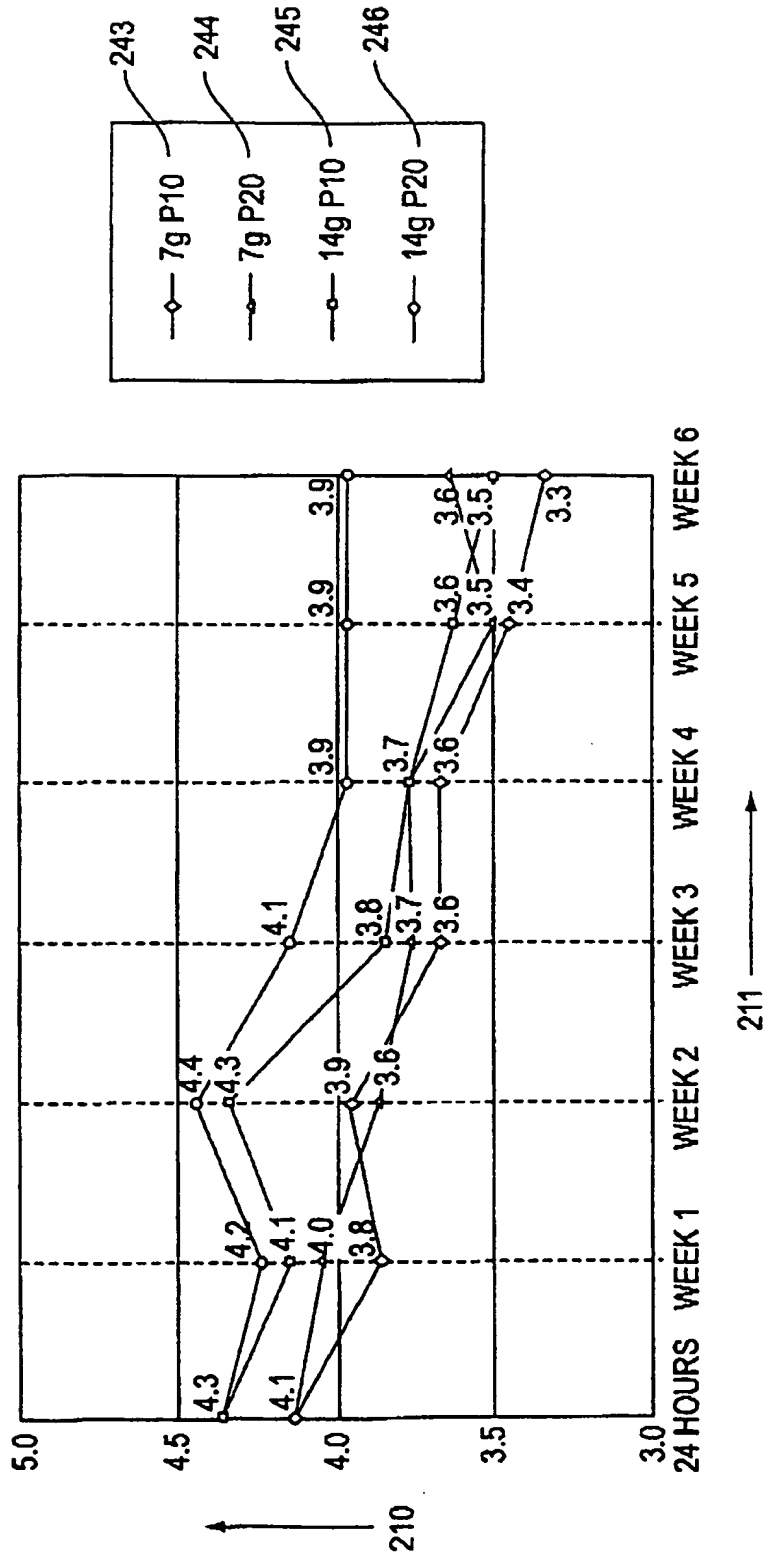


FS = FABRIC SOFTENER ONLY

-FS+S = NO FABRIC SOFTENER, SACHET ONLY

+FS+S = FABRIC SOFTENER AND SACHET

FIG. 14E



SCALE USED 5 = VERY STRONG TO 1 = VERY WEAK

FIG. 15

5 = VERY STRONG

4 = STRONG

3 = FAIR

2 = WEAK

1 = VERY WEAK

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/01789

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 A61L9/04 B65D75/30

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B65D A61L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 605 165 A (VAN LOVEREN AUGUSTINUS G ET AL) 12 August 1986 see column 8, line 43 - column 9, line 46; figures 1-17; examples ----	1-10
Y	US 4 521 541 A (RUTHERFORD HOWARD J ET AL) 4 June 1985 cited in the application see column 7, line 59 - column 13, line 29; figures ----	1-10
A	EP 0 081 791 A (HENKEL KGAA) 22 June 1983 see page 6, line 12 - page 8, line 10; figures -----	1-10



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

29 June 1999

Date of mailing of the international search report

07/07/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Olsson, B

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/01789

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 4605165 A	12-08-1986	NONE	
US 4521541 A	04-06-1985	US 4543367 A US 4542162 A	24-09-1985 17-09-1985
EP 0081791 A	22-06-1983	DE 3149508 A	23-06-1983

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.